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## THERMOLUMINESCENCE AND ANTARCTIC METEORITES

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**Abstract**

The level of natural thermoluminescence (TL) in meteorites is the result of competition between build-up, due to exposure to cosmic radiation, and thermal decay. Antarctic meteorites tend to have lower natural TL than non-Antarctic meteorites because of their generally larger terrestrial ages. However, since a few observed falls have low TL due to a recent heating event, such as passage within  $\sim 0.7$  astronomical units of the Sun, this could also be the case for some Antarctic meteorites. Dose rate variations due to shielding, heating during atmospheric passage and anomalous fading also cause natural TL variations, but the effects are either relatively small, occur infrequently, or can be experimentally circumvented.

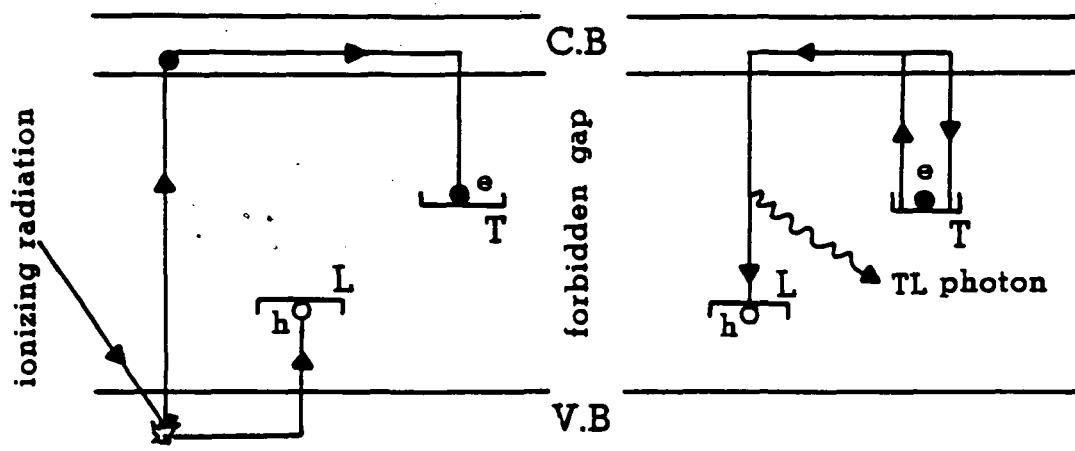
The TL sensitivity of meteorites (TL emitted after the natural TL has been removed and the sample administered a standard radiation dose) reflects the abundance and nature of the feldspar. Thus intense shock, which destroys feldspar, causes the TL sensitivity to decrease by 1-2 orders of magnitude, while metamorphism, which generates feldspar through the devitrification of glass, causes TL sensitivity to increase by a factor of  $\sim 10^5$ . The TL-metamorphism relationship is particularly strong for the lowest levels of metamorphism. The order-disorder transformation in feldspar also affects the TL emission characteristics and thus TL provides a means of paleothermometry.

**1. Introduction**

Measurement of the thermoluminescence (TL) properties of meteorites provides certain clues into diverse aspects of their history and that of the ice on which they are found. The present article describes the theoretical and experimental background to TL measurement of meteorites and summarizes current understanding of the factors which determine their TL properties. It ends with a summary of the main points with particular reference to the value of including TL in the routine preliminary survey of Antarctic meteorites.

**2. Theory**

As first pointed out by Randall and Wilkins (1945) and Garlick and Gibson (1948), the band model provides a reasonable framework for understanding TL in semi-conductors, although for most phosphors details are known only in a fragmentary way (Fig. 1). Any ionizing radiation promotes electrons from the valence band (V.B) to the normally unoccupied conduction band (C.B) where they pass freely through the lattice. Located through most crystals are intrinsic and impurity produced defects which provide sites (e.g. T and L) for metastably



**Figure 1:** Band model for a semi-conductor crystal lattice explaining the main features of thermoluminescence. Ionizing radiation excites an electron from the valence band (V.B) to the conduction band (C.B) where it moves through the lattice until trapped at a site (T)  $\sim 1$  eV below the C.B. Thermal stimulation enables the trapped electron to escape the trap and return to the ground state via a luminescence center (L) at which it releases some of the excitation energy in the form of a visible photon. The process may also involve recombination of the electron and the positive hole that was created by ionization (h).

trapping the excited electron at various energies, but typically  $\sim 1$  eV, below the conduction band. The rate of trap filling can be represented by a relationship such as

$$\frac{dn}{dt} = \alpha R(N-n) \quad (1)$$

where  $n$  is the number of trapped electrons,  $N$  is the number of available traps,  $R$  the dose rate,  $t$  is time and  $\alpha$  a rate constant. For a meteorite the major sources of ionizing radiation are primary and secondary cosmic rays for which dose rates are thought to be on the order of  $\sim 1$  rad/year.

The trap depth constitutes an activation barrier to decay of excited electrons which can be overcome by heating the sample to the appropriate temperature. Once released from the traps the electrons may become retrapped at the same or other defects, or fall back to the valence band by a number of pathways. It is those which involve radiative transitions at visible wavelengths that are responsible for the TL phenomenon. This part of the TL mechanism can also be modeled quantitatively. Assuming first order decay kinetics, the rate of decay of excited electrons is given by

$$\frac{dn}{dt} = -\beta n \quad (2)$$

where  $\beta$  is the first-order decay constant. The relationship between  $\beta$  and the trap depth ( $E$ ) and temperature ( $T$ ) is given by the Arrhenius equation so that

$$\frac{dn}{dt} = -Sn \exp(-E/kT) \quad (3)$$

where  $S$  is the Arrhenius or pre-exponential factor, and  $k$  is the Boltzman constant.

Cosmic ray exposure ages for meteorites are  $\sim 10^7$  year, so that equilibrium will be achieved for the electrons in most traps. Setting the rate of decay equal to the rate of build-up, and re-arranging, the relationship between number of trapped electrons, and therefore the level of TL, and the dose-rate and ambient temperature is given by

$$n = \frac{N}{1 + [S/\alpha R \exp(-E/kT)]} \quad (4)$$

There are many such quantitative models for the TL process, most of greater sophistication, and it is not clear which is applicable to meteorites. There is strong evidence that the decay kinetics of the TL or ordinary chondrites is second order (McKeever, 1980). However, all models predict that the level of TL naturally present in a meteorite will normally be at an equilibrium and that the level will be related directly to dose rate and reciprocal temperature. It is also possible, in principle, for the TL level to be out of equilibrium if the traps are all filled or if insufficient time has elapsed since the system was last drained for equilibrium to have been re-established. Insertion of plausible values into equations 1 and 3 suggests that it may take  $\sim 10^5$  y to achieve equilibrium, so that it may be feasible to empirically determine the time taken to reach TL equilibrium by looking for a relationship between natural TL and exposure age for the dozen or so meteorites with extremely short exposure ages.

Thermoluminescence can be induced in the sample after the natural TL has been removed, by, for instance, momentary heating to  $\sim 500^\circ\text{C}$ , by irradiation in the laboratory. The level of induced TL depends on the number and type of traps, and the details of the induced TL emission depend on such factors as the crystallography of the phosphor.

### 3. Apparatus and Procedures

Samples are heated on a nichrome strip in a nitrogen atmosphere at carefully controlled linear heating rates of  $5\text{--}20^\circ\text{C}/\text{sec}$ ; a signal from a chromel-alumel thermocouple placed under the center of the heating strip provides a temperature feedback to the control unit (Fig. 2). The emitted TL is detected with a specially designed electronically and magnetically shielded photomultiplier tube behind a heat filter and a blue filter (Mills et al., 1981). The blue filter and an aperture keep to a minimum the red black-body radiation from sample and heating strip. Rate counting electronics provide a high signal-to-noise input for storage by a microprocessor or feeding directly to the Y-axis of an X-Y plotter with temperature on the X-axis. The resulting plot of TL against temperature, termed a glow curve, contains a number of peaks corresponding to the number of discrete trap levels. For natural meteorite samples, there is a relatively sharp peak at  $\sim 250^\circ\text{C}$  and a broader peak at  $\sim 450^\circ\text{C}$ . For the induced TL there is a broad peak between  $100\text{--}300^\circ\text{C}$ . In fact, all the "peaks" referred to are, in fact, composites of many smaller peaks.

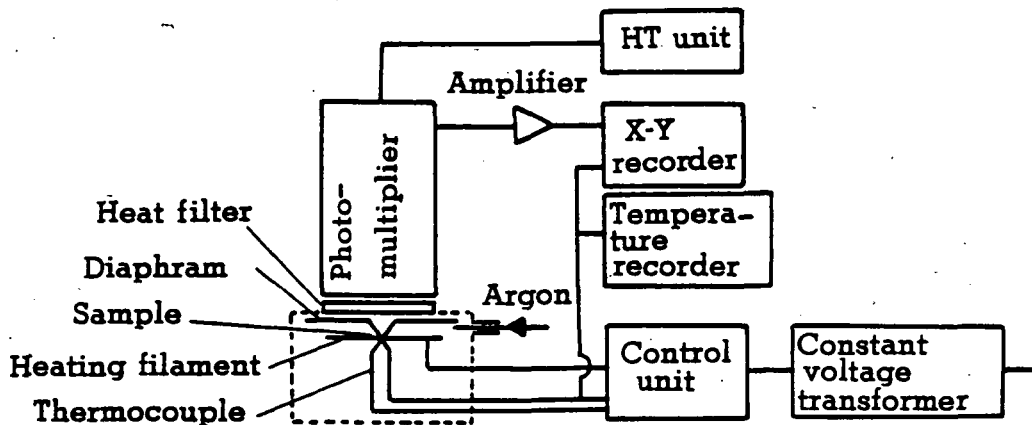


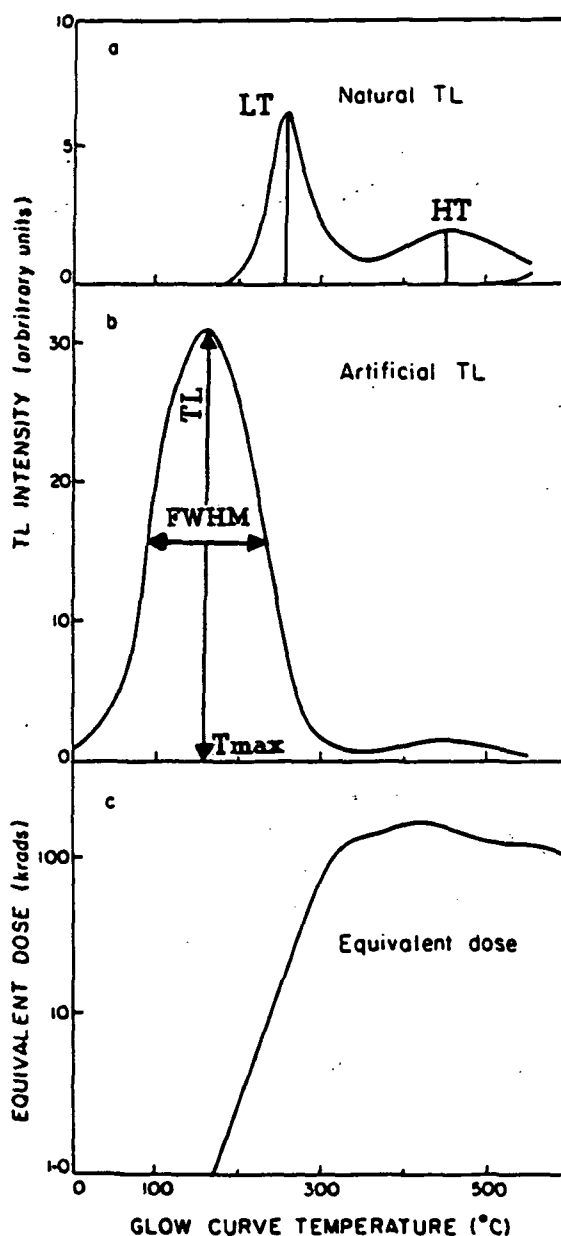
Figure 2: Schematic diagram of apparatus for the measurement of thermoluminescence. The function of the recorders and the control units are often performed by a microprocessor.

Samples are prepared either as powders by removal of the metal with a hand magnet and grinding so as to pass through a 100 mesh sieve, or as  $\sim 1 \mu\text{m}$  grains deposited from an acetone suspension into a 1 cm Cu disks. The powders are placed in Cu pans for TL measurement, whereas the disks can be measured directly and are in a convenient form for storage. Typically each sample is measured in duplicate and the TL of each duplicate measured 4-6 times; the natural TL is first measured, and thus drained, and then the sample irradiated with a Co-60 or Sr-90 source (typically to 25 krad) and the induced TL measured 3-5 times, depending on reproducibility. At the beginning and end of a days measurements, samples of the Dhajala meteorite are run as a normalization standard. Dhajala has a TL sensitivity in the middle of the observed range and is available in reasonable quantities. Its use as a standard enables interlaboratory comparison of data.

#### 4. Data Reduction

The information available from the measurement of natural TL and from the induced TL is very different and this is reflected in the means of data reduction (Fig. 3). The main object in measurement of natural TL is to assess the level of natural TL in a way which removes the effects due to variations not related to R and T, such as sample heterogeneity or considerable variations in the number of available traps. The two methods available are (1) to measure the ratio of the low temperature peak (LT) to the high temperature peak (HT) (Fig. 3a), and (2) to measure the ratio of the natural TL to the induced TL at a given glow curve temperature; this ratio can be multiplied by the test dose and resulting quantity has the units of dose and is termed the "equivalent dose". The first method presupposes that HT and LT are equally sensitive to the interfering effects, and the second is subject to greater experimental errors such as thermal lag between sample and heating strip (Sears and McKeever, 1980). A plot of equivalent dose against glow curve temperature permits a convenient means of examining the extent of thermal draining (Fig. 3c).

Figure 3: Methods of data reduction for (a) natural TL and (b) induced TL (artificial TL). (c) The rates of the curves for natural and induced TL (artificial TL) (after Melcher, 1981a).



Three measurements may be usefully made from the glow curve for the induced TL: 1) the height of the peak at maximum emission, which is referred to as TL sensitivity and is frequently normalized to that of the Dhajala meteorite; 2) the width of the peak at half maximum (FWHM or simply peak width); 3) the temperature of maximum TL emission (or peak temperature) (Fig. 3b).

## 5. Natural TL Levels in Meteorites

The levels of natural TL in a number of ordinary chondrites is shown in histogram form in Fig. 4. Slightly different distributions are displayed by the observed falls and the finds. The falls have preferred values between 2 and 7,

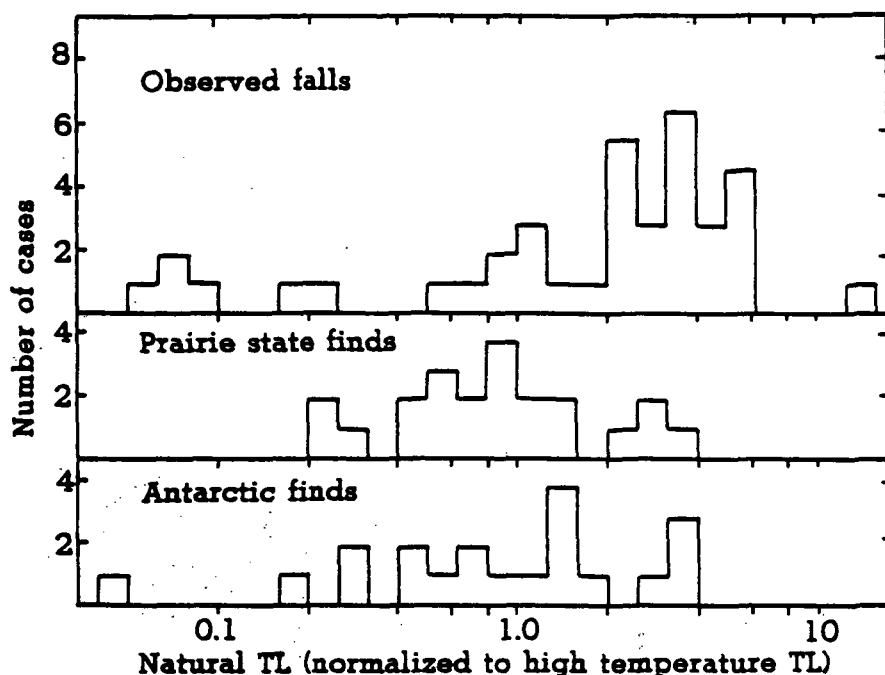


Figure 4: Histograms of the natural TL levels in meteorites that were observed to fall within the last 250 years (top, McKeever and Sears, 1980), meteorites that were found in the Prairie States of the U.S.A. (middle, Sears and Durrani, 1980) and those that were found in the Antarctic (bottom, present study).

but spread over 2 orders of magnitude and extend down to peak height ratio values of less than 0.1. The Antarctic and Prairie State finds also show considerable spread which is similar for the two sources and skewed to lower values than for the falls ( $\sim 0.2$  to  $\sim 3$ ) (Sears and Durrani, 1980; McKeever and Sears, 1980).

The difference in TL between the falls and finds, which is an order of magnitude, is probably associated with differences in terrestrial age. The Al-26 activity of Antarctic meteorites is about 20% lower than non-Antarctic meteorites (Fig. 5) and since there is no evidence from other cosmogenic isotopes that Antarctic meteorites have suffered significantly different exposure histories, this must reflect terrestrial ages which are a significant fraction of the half-life of Al-26 (Evans et al., 1982). The extensive measurements, based on other cosmogenic nuclides, by Nishiizumi et al. (1979, 1981) have shown that the terrestrial ages of Antarctic meteorites are often  $\sim 10^5$  y. Theoretically, a relationship between natural TL and terrestrial age is to be expected, since the cosmic ray dose rate decreases and the ambient temperature increases, after a meteorite falls to earth. The equilibrium TL level has therefore to adjust to a new low level. Fig. 6a shows the decrease in natural TL as a function of terrestrial age, and Fig. 6b shows the abundance of two cosmogenic isotopes in relation to their natural TL levels (Sears and Durrani, 1980; Melcher, 1981a). In both cases there is a suggestion that the natural TL is falling to lower levels as the terrestrial ages of the meteorites increases.

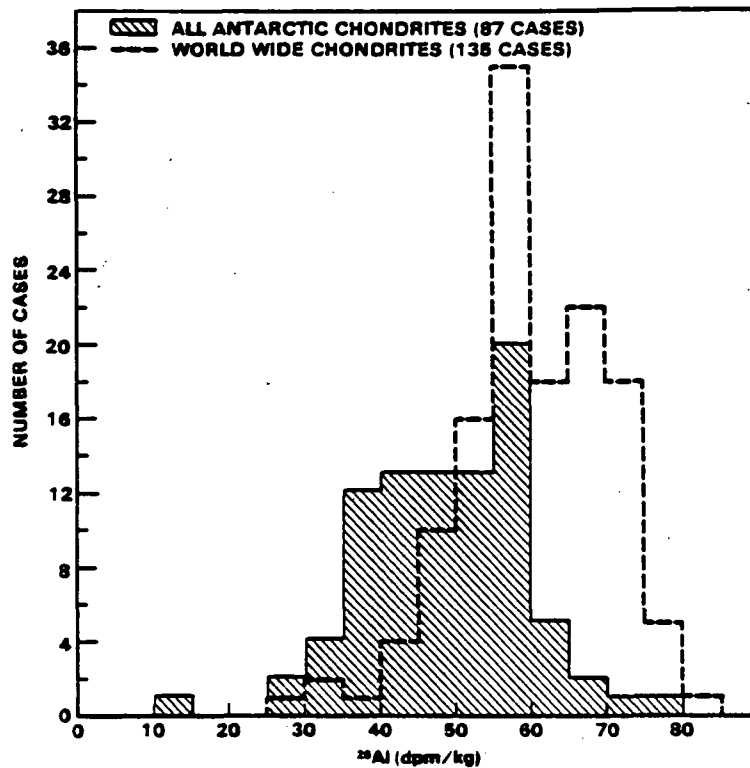


Figure 5: Histograms of the Al-26 content of non-Antarctic and Antarctic meteorites (from Evans et al., 1982).

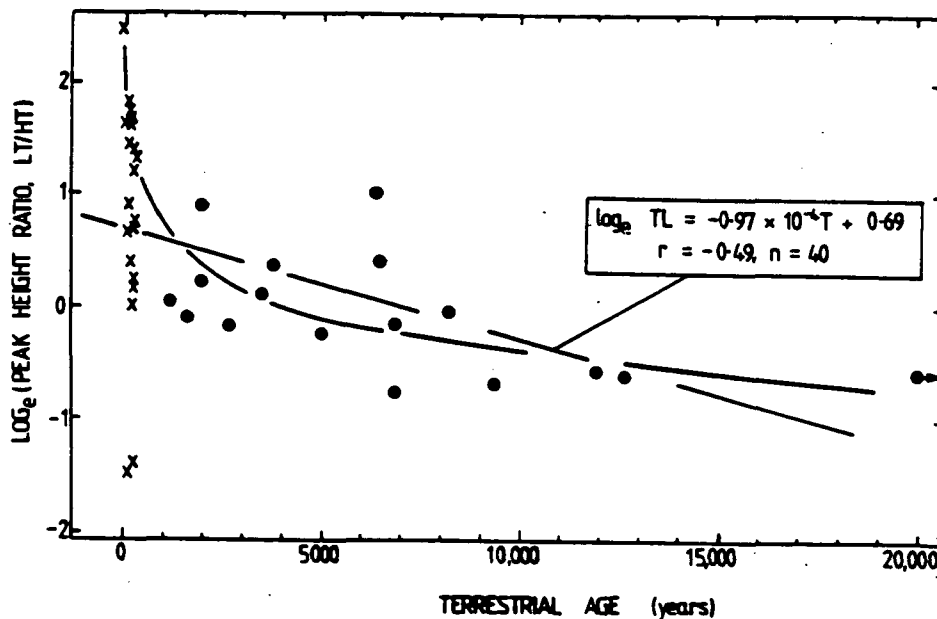


Figure 6a: Plot of natural TL against terrestrial age for observed falls (crosses) and finds whose terrestrial age has been determined from their C-14 contents (dots) (from Sears and Durrani, 1980). The straight line is a least squares fit, the regression equation and parameters are given in the box, assuming a first-order decay process equation (2), while the curve was drawn in by eye to suggest the form expected from second-order decay.

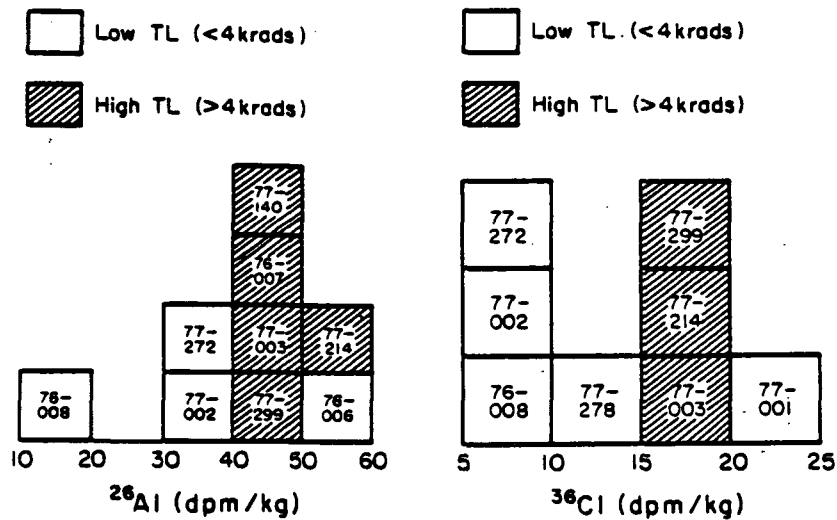


Figure 6b: Histograms of Al-26 and Cl-36 content of Antarctic meteorites of known natural TL levels (in equivalent dose in rad) (from Melcher, 1981a).

Prairie Network meteorites appear to have terrestrial ages within a few half-lives of carbon-14 (5220 y) (Boeckl, 1972), yet display a similar natural TL spread to the Antarctic meteorites which have terrestrial ages comparable with the half-life of Al-26 (750,000 y). The similarity in the TL despite different terrestrial ages presumably reflects the higher storage temperature for the Prairie Network meteorites.

A process other than large terrestrial age is involved in determining the natural TL levels in certain meteorites, since several meteorites observed to fall within the last 200 y have TL levels of  $\sim 0.1$  (Fig. 6a). Such meteorites have experienced a thermal and/or radiation history very different to the others, for instance they may have been reheated recently so that their TL was drained and unable to return to the equilibrium value before fall to earth. The lack of low temperature TL, but apparently normal levels of high temperature TL, is most evident on a plot of equivalent dose against glow temperature (Fig. 7) (Melcher, 1981b). Solar heating during close passage to the Sun provides a reasonable explanation of these data. The temperature (T) of an object at astronomical units (a.u.) from the Sun is given by

$$T = \frac{CB}{d^2 S \delta} \gamma \quad (5)$$

where C is the solar constant, B the cross section and S the surface area of the object,  $\delta$  is Stefan's constant and  $\gamma$  the ratio of the absorbed and emitted radiation (typically  $\sim 0.5$ ). Assuming plausible figures this becomes

$$T = 278 \left( \frac{\gamma}{d^2} \right)^{1/4} \quad (6)$$

The important orbital parameter as far as TL is concerned is the perihelion distance. For the Pribram and Lost City meteorites whose fireballs were photographed at several locations, the perihelia were 0.790 and 0.967 which correspond to temperatures of 255 and 236 K, respectively. One of the fireballs



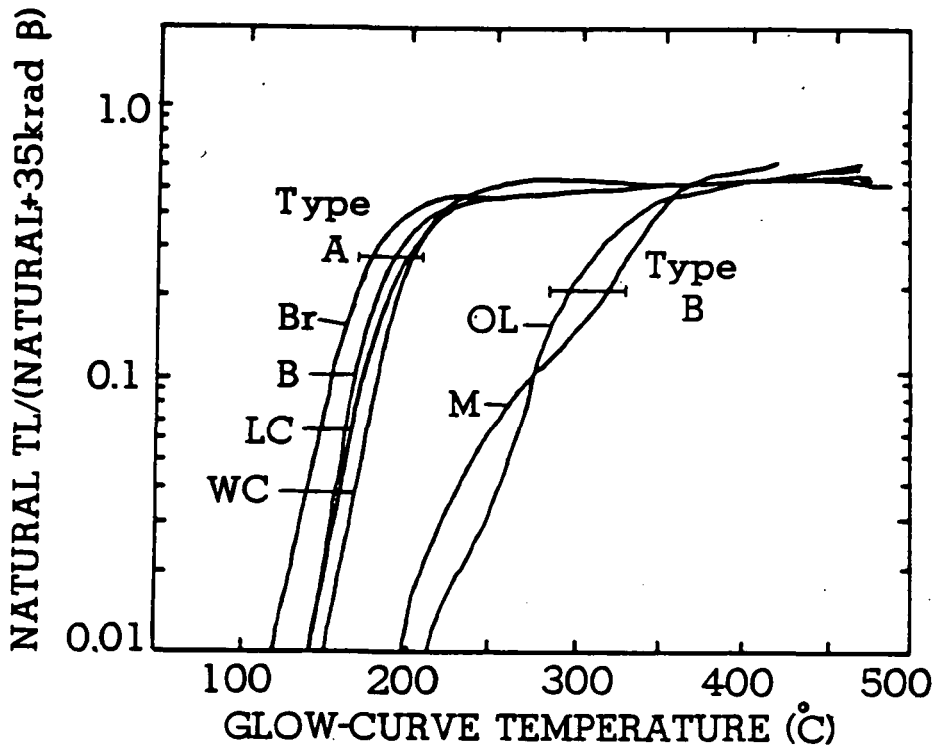


Figure 7: Plots of equivalent dose (natural TL/induced TL times test dose) against glow curve temperature for several observed falls. Most meteorites (type A) have no natural TL <200°C in the glow curve due to thermal drainage, probably under equilibrium conditions in space. However, a few (type B) have suffered unusually high drainage, perhaps due to close solar passage (from Melcher 1981a, McKeever and Sears, 1980).

observed by the Prairie Network, but which did not yield a meteorite, was Meteor 40503 with a perihelion of 0.722 and corresponding temperature of 275 K. On the basis of the above theory if we assume

$$n \propto \exp (E/kT) \quad (7)$$

then the ratio of trapped electrons for a meteorite with perihelion at 0.722 and 0.967 a.u. is very approximately given by

$$\begin{aligned} \frac{n_{0.722}}{n_{0.967}} &= \exp \left( \frac{E}{k275} - \frac{E}{k236} \right) \\ &= 0.01 \end{aligned} \quad (8)$$

for a trap depth of ~1 eV. A two-order of magnitude difference in natural TL is consistent with known variations in perihelia and plausible TL kinetic models. In an attempt to test this idea, McKeever and Sears (1980) and Melcher (1981b) compared the natural TL variations of falls with  $^3\text{He}/^{21}\text{Ne}$  ratios, a ratio less than 0.3 is an indication of gas-loss due to solar heating. However, the

meteorites which had suffered gas-loss had normal TL suggesting that if heating was involved that it occurred long enough ago that the TL had recovered. A more reasonable test of the idea is to examine the natural TL of meteorites known to have small perihelia when they entered the atmosphere.

There are other instances in which dose-rate or temperature changes can occur which influence natural TL levels. Variation in the dose-rate may occur with the attenuation of primary cosmic radiation and build-up of secondary radiation. The natural TL level is 10-15% higher at the center of a 55 x 40 cm slab of the Estacado meteorite (Fig. 8a) (Sears, 1975) and a 50% variation in

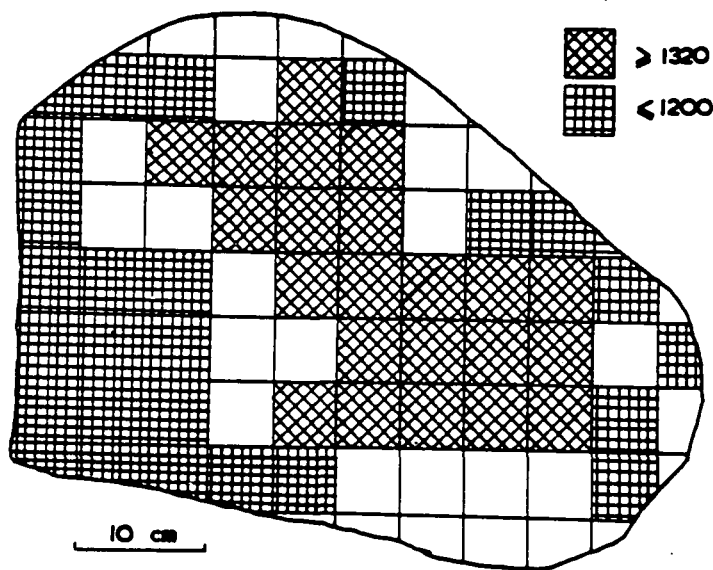


Figure 8a: Natural TL in a 40 x 55 cm slab of the Estacado meteorite (from Sears, 1975).

the TL of the Lost City meteorite was observed over a 10 cm distance (Fig. 8b) (Vaz, 1971), both most plausibly caused by cosmogenic dose rate variations. Factors of 2 change in natural TL also occur in two core samples taken from the St. Severin meteorite by Lalou et al. (1970) (Fig. 8c). As yet, however, there is no indication that shielding effects can result in order of magnitude natural TL variations, and, at most probably only contributes to the scatter in Fig. 4. The thermal pulse caused by atmospheric passage drains the TL over the outermost 5 mm or so (Fig. 8b), but this effect can easily be avoided by careful sampling.

A mechanism also exists in which natural TL decays in a poorly understood but extremely rapid means in certain materials. Hasan et al. (1985) found that certain shergottites and the Kapoeta howardite displayed such "anomalous fading" and suggested that it was only affecting samples in which the feldspar is present in the low temperature form. If this is the case, then anomalous fading will not generally be a problem since such meteorites are rare, and, in any event, seems to be readily detected by laboratory tests. McKeever and Durrani (1980) found no evidence for anomalous fading in two ordinary chondrites, Barwell and Saratov.

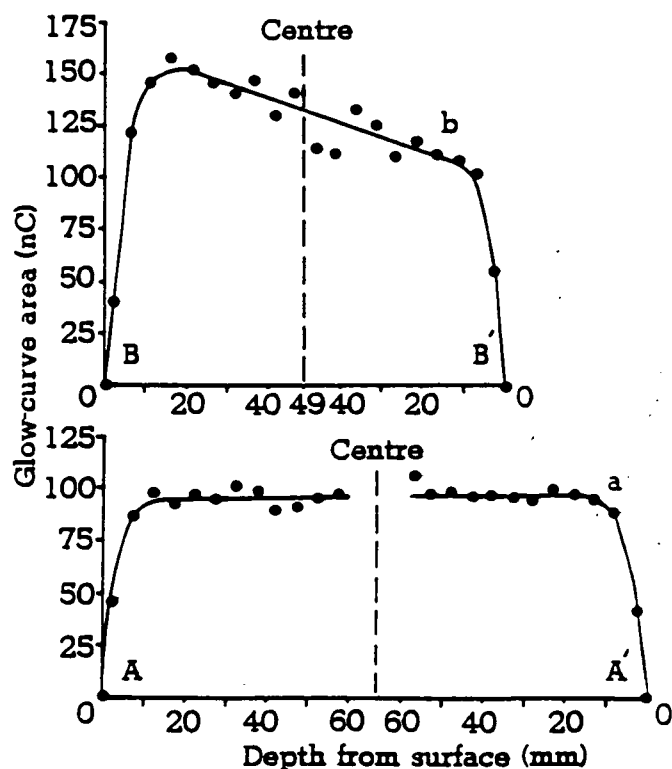


Figure 8b: Natural TL variations in two bars removed from the Ucera and Lost City meteorites. The outer 5 mm or so of each bar also shows draining of the TL due to the heat generated by atmospheric passage (from Vaz, 1971).

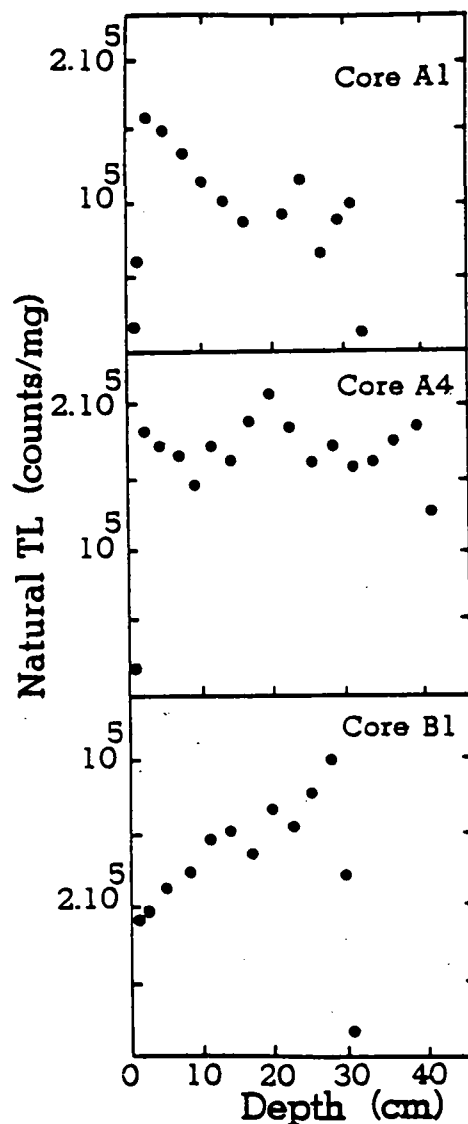


Figure 8c: Natural TL along three cores taken from two fragments (A and B) of the St. Severin chondrite. Cores A1 and A2 were approximately perpendicular (for the precise orientations, see Lalou et al., 1970, from which the data were taken).

## 6. The Induced TL Properties of Meteorites

The TL sensitivity of meteorites shows a  $10^6$ -fold range, the ordinary chondrites alone show a  $10^5$ -fold range and the shergottites have values below them (Sears et al., 1980; Hasan and Sears, 1985). With a few exceptions, the mineral responsible for the TL in meteorites is feldspar and the range in TL sensitivity reflects the amount of the phosphor present. Heavily shocked ordinary chondrites, in which the feldspar has been fused or partially converted to maskelynite, have TL sensitivities one to two orders of magnitude lower than unshocked equilibrated chondrites. Such low TL sensitivities can be reproduced by annealing an unshocked chondrite to  $\sim 1100^\circ\text{C}$ , which is similar to the post-shock residual temperature for heavily shocked chondrites inferred from studies of the metal and sulfide (Fig. 9; Liener and Geiss, 1968; Sears et al., 1984).

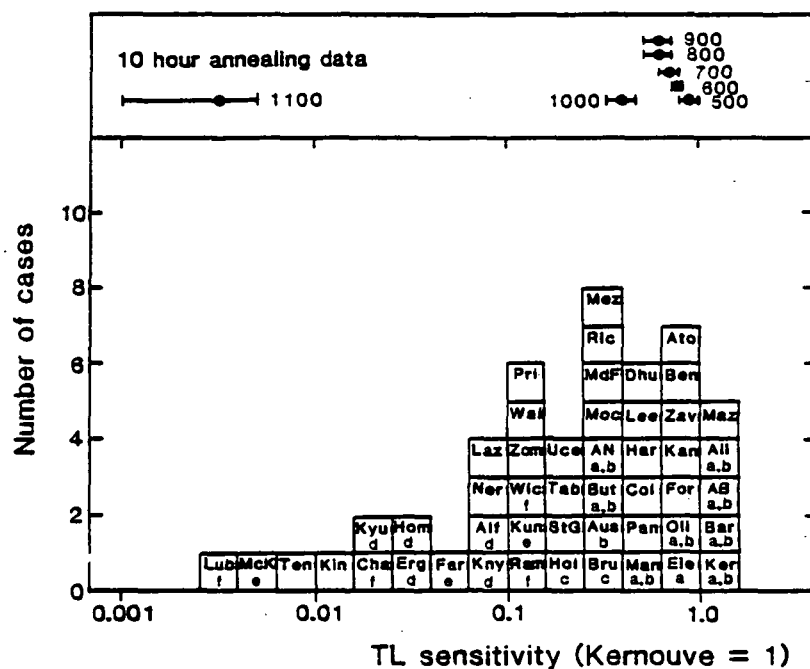


Figure 9: Histogram of TL sensitivity levels in equilibrated ordinary chondrites of a variety of shock histories. The meteorites are identified by the first three letters of their names (see Sears, 1980, for identification) and their shock classification is also indicated (a, unshocked; f, heavily shocked). The data across the top of the histogram indicate the TL sensitivity of Kernouve samples which have been annealed for 10 h at the temperatures indicated (in  $^\circ\text{C}$ ) (Sears et al., 1984).

Unshocked ordinary chondrites display a large range of TL sensitivity which is metamorphism-related (Fig. 10). The feldspathic constituent of unmetamorphosed ordinary chondrites is present as a glass which has little or no TL sensitivity. During relatively low-grade metamorphism the glass produces crystalline feldspar and the TL sensitivity increases accordingly. This has permitted a reliable subdivision of the type 3 chondrites into types 3.0 to 3.9 and many textural, compositional and isotopic variations are related to these types. The mechanism for metamorphism-related increase in TL was recently confirmed by

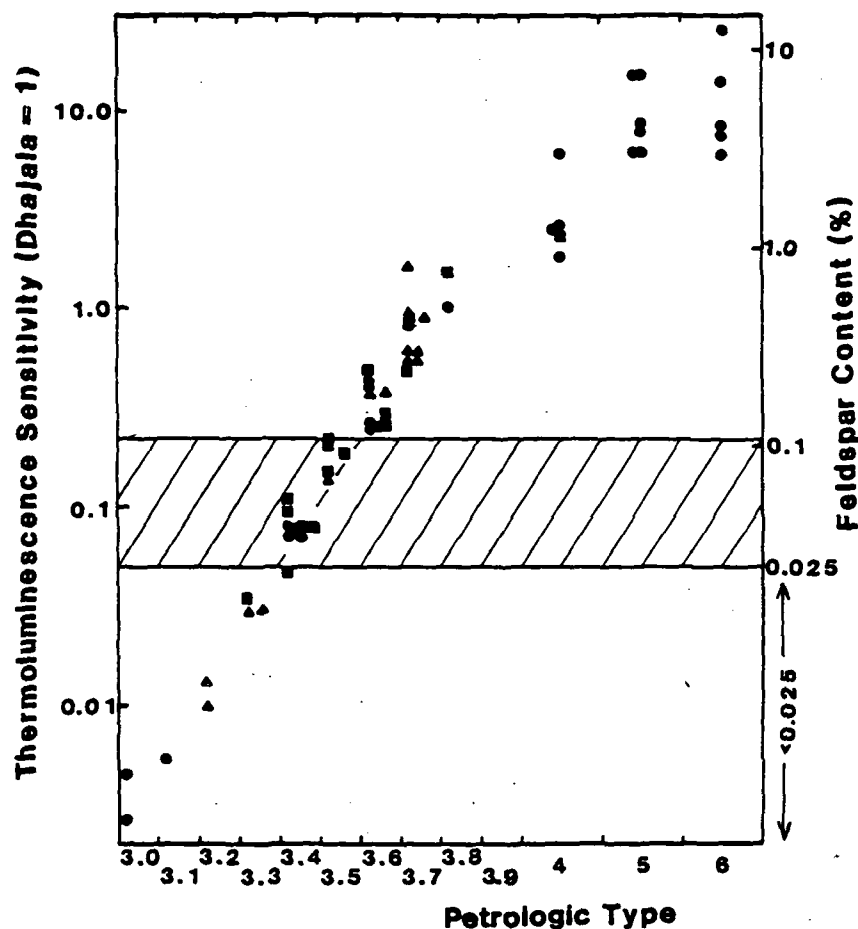


Figure 10: TL sensitivity variations in unshocked ordinary chondrites of a variety of petrologic types (from Guimon et al., 1985b). The cross-hatching refers to the TL sensitivity range in which the TL peak is narrow and at low glow-curve temperatures and probably indicates that the feldspar is in the low-temperature form. The right-hand axis is an estimate, based on the TL, of the feldspar content of the meteorites. The symbols refer to the literature sources for the data: circles, Sears et al. (1980); squares, Sears et al. (1982); triangles, Sears and Weeks (1983).

annealing a type 3.4 chondrite in a manner known to cause divitrification of synthetic glasses. A factor of 7 increase in the TL at 200°C in the glow curve resulted after only 200 h of annealing (Fig. 11). At metamorphic levels equivalent to a TL sensitivity of 0.025 (where Dhajala = 1) the induced TL curve consists of a narrow peak (100°C) at relatively low temperatures in the glow curve (~100°C), but at high levels of metamorphism the curve becomes broader (160°C) and moves to higher glow curve temperatures (200°C). This increase in TL peak temperature and peak width can be reproduced in the laboratory by annealing at >700°C for 100 h either a low petrologic type meteorite or a terrestrial feldspar in the low-temperature (ordered) form (Fig. 12). In the case of the terrestrial feldspar, the x-ray diffraction pattern indicated that the TL changes were associated with the onset of the conversion of the ordered to the disordered form (Guimon et al., 1984; 1985a).

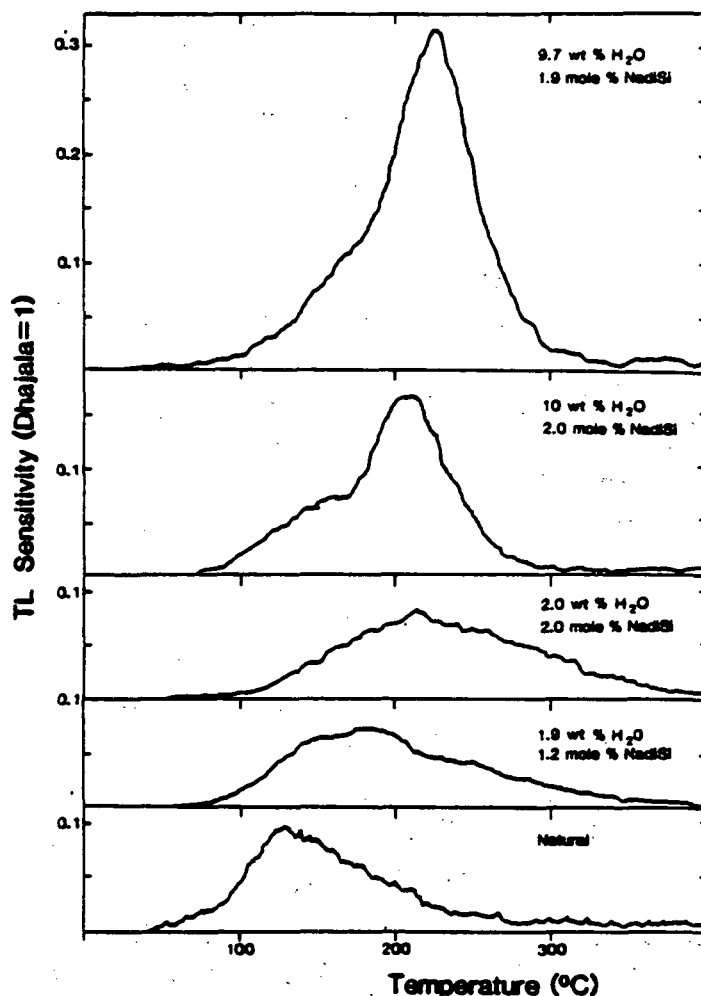


Figure 11: Glow curves for several samples of the Allan Hills A77011 chondrites which have been annealed at 755–850°C and 1 kbar for ~1 week with a variety of devitrification catalysts added (from Guimon et al., 1985b).

Although very different in origin, history, composition and petrology, the TL properties of the shergottites can be understood in very similar terms to those of the ordinary chondrites (Hasan and Sears, 1985). Mineral separation experiments indicate that the TL phosphor is also feldspar and the extensive maskelynitization suffered by the class is consistent with their very low TL sensitivity values. The shergottites also show systematic variation in peak temperature and width which can be understood in terms of feldspar being present in the high and low temperature forms in varying relative amounts (Fig. 13). Annealing treatments under much milder conditions than those used for ordinary chondrites produce spectacular TL sensitivity increases which is consistent with the known relative instability of the glass in shergottites relative to the igneous glass in type 3 ordinary chondrites (Fig. 14). Again, the temperature of the TL peak is related to the annealing temperatures.

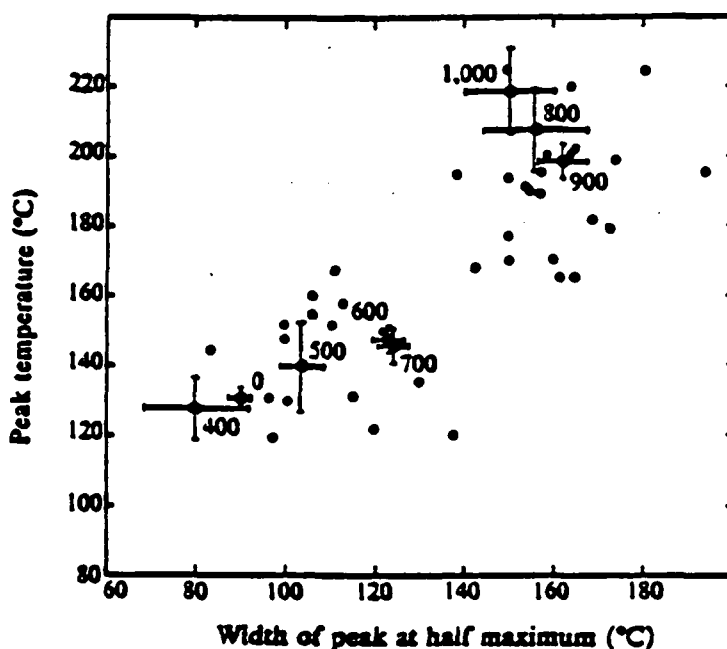


Figure 12: TL peak temperature against peak width for 39 type 3 ordinary chondrites (dots) and for a L3.4 chondrite (Allan Hills A77011) annealed for 200 h at the temperatures (°C) indicated (dots with error bars) (from Guimon et al., 1984).

## 7. TL as a Survey Technique for Antarctic Meteorites

The complexity and demands of surveying the Antarctic meteorites by TL are comparable with those of petrographic and electron microprobe surveys currently being performed. However, the nature of the available information is, by and large, very different. The following summarizes the above discussion with particular reference to the desirability of surveying recovered Antarctic meteorites with the technique and draws on the discussions published by Sutton and Walker (1985), Melcher (1981a) and McKeever (1982).

### Natural TL

1) Meteorites with high TL. None of the Antarctic meteorites yet measured have natural TL levels comparable with those of brightest observed falls. It seems fairly clear that a high natural TL can only reflect a recent fall, since the peak height ratio in even the most highly irradiated samples decays to  $\sim 1.0$  within a few thousand years (Fig. 6a).

2) Meteorites with low TL. Several processes can lead to a meteorite having abnormally low natural TL. The sample could have a particularly large terrestrial age, could have suffered reheating within the last  $10^5$  y or so, perhaps by having a small perihelion, or could be subject to anomalous fading. Anomalous fading can readily be identified by laboratory tests and seems to be relevant to relatively few meteorites.

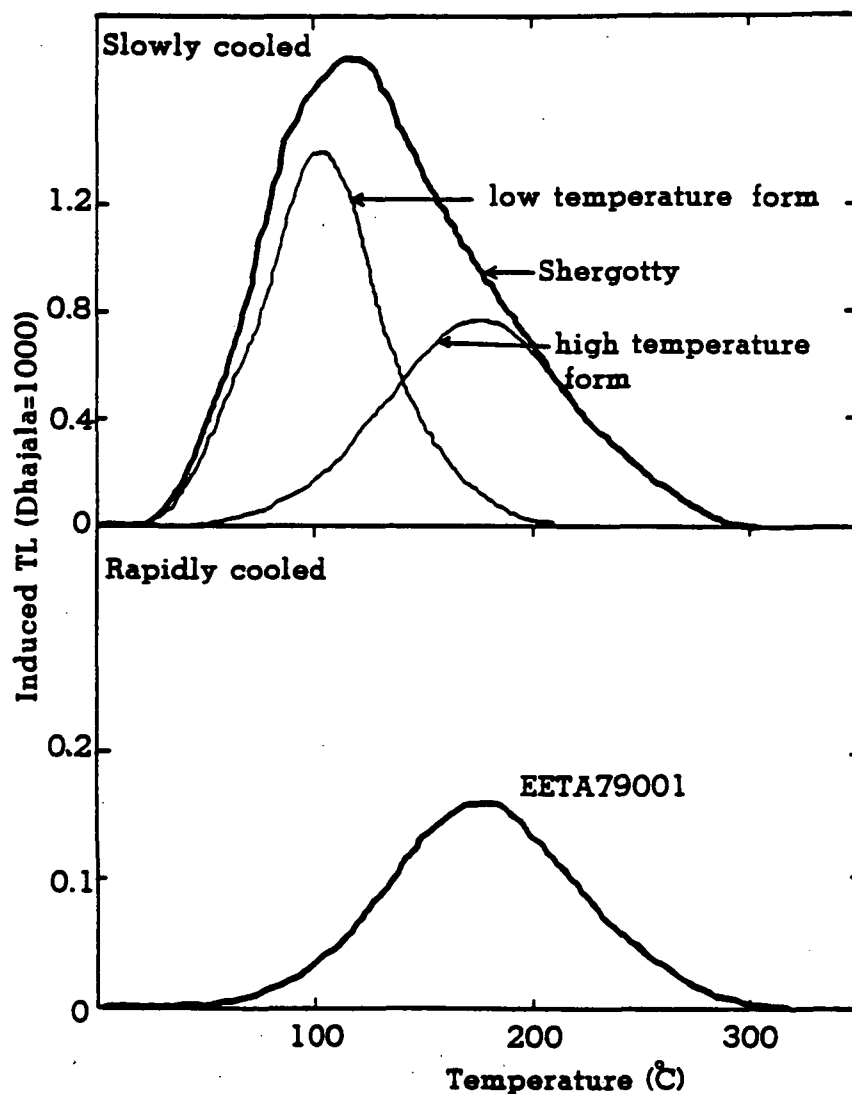


Figure 13: Glow curves for the Shergotty and Elephant Moraine A79001 shergottites with a suggested mechanism for explaining the peak temperature, peak width, and TL sensitivity. It is suggested that post-shock recrystallization of maskelynite in the meteorites produced feldspar in varying amounts and forms depending on cooling rate (Hasan et al., 1985).

3) Cosmogenic dose rate variations throughout the meteorite should cause variations in natural TL levels, but on the basis of existing data these seem to be relatively small. However, this point should be further tested by measurement of the natural TL variation throughout large meteorites.

4) Pairing is an important aspect of Antarctic meteorite curation and the problem could have serious implications for a variety of statistical studies. No single technique yet exists for unambiguous pairing of fragments of the same fall, but since natural TL levels span a range of over 100 and yet variations within a single meteorite seem to be less than ~50%, the natural TL may provide additional information on the question.



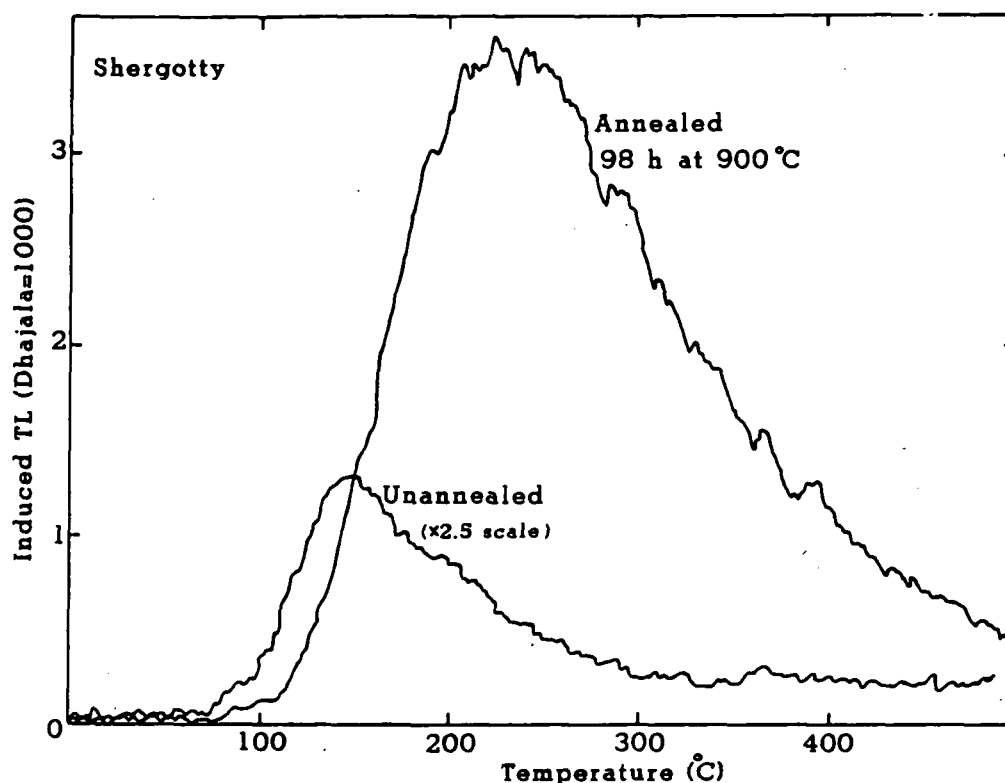


Figure 14: Glow curves for the Shergotty meteorite before and after an annealing treatment. The annealing produced considerable crystallization of the high temperature form of feldspar whose TL peak appears at 220°C in the glow curve (Hasan et al., 1985).

### Induced TL

- 1) Heavily shocked ordinary chondrites have TL sensitivities  $10^{-1}$  to  $10^{-2}$  times those of equilibrated unshocked ordinary chondrites.
- 2) While types 5 and 6 ordinary chondrites have similar TL sensitivities, type 4 chondrites tend to have TL sensitivities an order of magnitude or so lower than type 5,6 levels. Type 3 ordinary chondrites have TL sensitivities  $10^{-2}$  to  $10^{-5}$  times type 5,6 levels, and TL sensitivity provides a means of subdividing the type 3 chondrites according to their metamorphic history. All type 3 ordinary chondrites from the US Antarctic collection have been assigned to petrologic types on the basis of TL sensitivity.
- 3) Pairing studies are also possible using TL sensitivity, but the method is really only useful for type 3 chondrites which show a very large intra-chondrite spread. The pairing of several Allan Hills A77011 samples was discussed by Sears et al. (1982) who pointed out that the fragments paired on a petrographic basis had a rather large TL sensitivity spread and suggested that there may be fragments from two discrete falls present.

## Acknowledgements

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